Reciprocity relations in two-dimensional percolation theory

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The Dykhne method [Sov. Phys. JETP 32, 63, 348 (1971)] is used to analyze the relations between the effective characteristics of reciprocal systems that differ from each other in that the components interchange places. Reciprocity relations are obtained for a system in an external magnetic field, and also in the case when the phenomena that occur in the medium are described by several currents and fields. This permits the galvanomagnetic properties of two-component systems to be determined outside the region of the metal-insulator phase transition, as well as their thermoelectric properties in a zero magnetic field at equal concentrations of the components.

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1. Owing to the considerable difficulties that arise in the theoretical analysis of various properties (e.g., conductivity) of inhomogeneous media, each exact analytic solution takes on a special role. In this respect, two-dimensional systems (d = 2, where d is thedimensionality of the space) are in a more favorable position than three-dimensional ones, since a number of exact results for two-component media are available at d=2 (see Refs. 1 and 2, as well as 3-5). This is attained because in the two-dimensional case the dc equations admit of a symmetry transformation¹ from the initial system into a "reciprocal" one differing from the original by the substitution $\sigma_1 \neq \sigma_2$. Here σ_i is the conductivity of the *i*-th component. A relation, called hereafter the reciprocity relation,¹ can be established between the effective conductivities of the initial and of the reciprocal systems.

Reciprocity relations can be derived also in other more complicated cases (see Ref. 2 and the exposition below), so that a number of new results can be obtained for a reciprocal system if the properties of the initial one are known. They can be used also as checks and auxiliaries for various calculations. The most interesting consequences of reciprocity relations can be obtained, however, for those systems whose reciprocals go over into the initial ones following the additional substitution p - 1 - p; here p is the concentration of one of the component, for the sake of argument of the first ("metallic"). These include primarily randomly inhomogeneous media, which are attracting the greatest attention,⁶ as well as certain periodic media, such as of the checkerboard type.⁵ At $p = \frac{1}{2} [\tau = 0$, where τ $=(p - p_c)/p_c$ and $p_c = \frac{1}{2}$, these relations make it possible to determine for such systems the properties of the medium at the point of the metal-insulator (MI) transition.^{1,2} The significance of the reciprocity relations for such systems goes much farther, however, since they make it possible to relate the properties of the "metallic" ($\tau > 0$) and "insulating" ($\tau < 0$) phases.

The use of the reciprocity relation in the simplest form [see Eq. (8)] has enabled Dykhne¹ to prove the presence of a metal-insulator phase transition in a system and to obtain the critical concentration $p_c = \frac{1}{2}$. The same relation made it possible to obtain an independent proof⁵ of the relation between the critical exponents.⁶ The use of the Dykhne relation² between the components of the effective conductivity tensor in a weak magnetic field has enabled Shklovskii³ to find the behavior of the Hall coefficient in the entire range of the concentrations.

In the present paper the Dykne method^{1,2} is used to obtain reciprocity relations for an isotropic two-component system in a transverse magnetic field H. We consider also the case when the phenomena that occur in a medium are characterized by a matrix of kinetic coefficients, i.e., by a set of *n* currents and *n* fields, and the corresponding reciprocity relations are obtained. A similar problem arises in the study of the thermoelectric, thermal-diffusion, and other phenomena. We consider some consequences of the reciprocity relations, which make it possible, in particular, to find the galvanomagnetic properties of two-component systems outside the metal-insulator phase transition region and the thermoelectric properties at equal concentrations of the components at H=0.

2. We consider a two-dimensional two-component medium with an isotropic local conductivity $\sigma = \sigma(\mathbf{r})$. Following Dykhne,¹ we transform to a primed reference frame of the current density **j** and of the electric field E in accord with

$$\mathbf{j} = \lambda[\mathbf{n} \times \mathbf{E}'], \ \mathbf{E} = \lambda^{-1}[\mathbf{n} \times \mathbf{j}']; \tag{1}$$

here n is a unit vector normal to the (x, y) plane of the system, and λ is a certain constant independent of the coordinates. Under the transformation (1), the dc equations retain their form, and the conductivity in the primed frame is equal to

$$\sigma'(\mathbf{r}) = \lambda^2 / \sigma(\mathbf{r}) \,. \tag{2}$$

We note that the existence of the transformation (1) is due exactly to the fact that the system is two-dimensional, for in this case the equation curl E = 0 has only one component. On going to the primed system with the aid of (1), the equation divj = 0 is transformed into curl E' = 0 while curl E = 0 is transformed into divj'= 0.

At $\lambda^2 = \sigma_1 \sigma_2$ the primed system differs from the initial one by the substitution $\sigma_1 \neq \sigma_2$. We call such systems reciprocal. Following Dykhne¹ we obtain the reciprocity relation in the considered simplest case

$$\sigma_{e}(p)\overline{\sigma}_{s}(p) = \sigma_{1}\sigma_{2}.$$
(3)

The tilde marks here and below quantities pertaining to

the reciprocal system; p is the concentration of the first component. Relation (3) is quite general, being valid for any concentration p and for any form of the inclusions and their distribution.

Relation (3) can be useful in various approximate calculations. Thus, if σ_e is sought as a series in the powers of the second-component concentration c = 1 - p:

$$\sigma_{e} = \sigma_{i} (1 + cA_{i} + c^{2}A_{2} + \ldots), \quad A_{i} = A_{i} (\sigma_{2}/\sigma_{i}), \quad (4)$$

then (3) leads to relations between the coefficients A_i and \bar{A}_i :

$$A_1 + \widetilde{A}_1 = 0, \quad A_2 + \widetilde{A}_2 = A_1^2, \ldots, \quad \widetilde{A}_i = A_i(\sigma_1/\sigma_2).$$
(5)

For circular inclusions it is easily obtained, by a known method, 7

$$A_{i} = -2(\sigma_{i} - \sigma_{2})/(\sigma_{i} + \sigma_{2}), \qquad (6)$$

so that the first equation of (5) is automatically satisfied. The equations in (5) are valid also for inclusion of arbitrary shape and of arbitrary distribution.

For randomly inhomogeneous media and for certain periodic models (e.g., of the checkerboard type), the additional substitution p - 1 - p transforms the reciprocal system into the initial one:

$$\tilde{\sigma}_e(1-p) = \sigma_e(p). \tag{7}$$

The condition (7) imposes rather stringent limitations on the statistical properties of the system, so that the relation (8), which follows from (3) and (7), is less general than (3). We note that for a randomly inhomogeneous medium Eq. (7) is valid also in the three-dimensional case. In particular, it is satisfied by the approximate expressions obtained for σ_e by the method of the effective-medium theory (EMT),⁸ both at d=2and at d=3. When the equations of Ref. 8 are used for σ_e it must be taken into account that the continuous case considered here corresponds in Ref. 8 to z=6 nearest neighbors at d=3 and to z=4 at d=2.

When condition (7) is satisfied, Eq. (3) takes the form $^{1}\,$

$$\sigma_{e}(p)\sigma_{e}(1-p) = \sigma_{1}\sigma_{2}. \tag{8}$$

Equation (8) was used by Dykhne¹ to prove the existence of a metal-insulator transition at $\sigma_2 = 0$ and to find the critical concentration $p_c = \frac{1}{2}$. It can be used also for an independent proof⁵ of the connection between the critical exponents.⁶ Moreover, relation (8) makes it possible to determine the conductivity of the system in the entire $p < \frac{1}{2}$ region if σ_e is known for all $p > \frac{1}{2}$ (or vice versa).

It is curious to note that the expression obtained for σ_e by the EMT method⁸ (d=2)

$$\sigma_{e} = (p^{-1}/_{2}) (\sigma_{i} - \sigma_{2}) + [(p^{-1}/_{2})^{2} (\sigma_{i} - \sigma_{2})^{2} + \sigma_{i} \sigma_{2}]^{\frac{1}{2}}, \qquad (9)$$

satisfies Eq. (8). Equation (9) therefore gives not only the correct answer for the critical concentration $p_c = \frac{1}{2}$, but also the exact result that follows from (8) at $p = \frac{1}{2}$ (Ref. 1):

$$\sigma_e = (\sigma_1 \sigma_2)^{\frac{1}{2}}.$$
 (10)

This is in fact the reason why the EMT is apparently a rather successful approximation in the two-dimensional case (especially at $\sigma_2 \neq 0$ -Ref. 8).

Expressing (8) in the form $\sigma_e(\tau)\sigma_e(-\tau) = \sigma_1\sigma_2$, where $\tau = (p - p_c)/p_c$ is the parameter of the proximity to the transition in terms of the concentration, and proposing that $\sigma_e(\tau)$ can be expanded in powers of τ , we obtain the relations between the various derivatives of $\sigma_e(\tau)$ at the point $\tau = 0$. In particular,

$$\sigma_{e}''(0) = \sigma_{e'}(0) / \sigma_{e}(0).$$
(11)

It follows therefore that if $\sigma'_e(0) \neq 0$ we have $\sigma''_e(0) > 0$, i.e., the $\sigma_e = \sigma_e(\tau)$ curve is concave upward at the point $\tau = 0$. On the other hand if $\sigma'_e(0) = 0$ we also have $\sigma''_e(0) = 0$ so that $\tau = 0$ is an inflection point.

From (1) follow directly reciprocity relations for the relative quadratic fluctuations of the current density Δ_j and of the electric field Δ_E :

$$\Delta_j = \bar{\Delta}_E, \quad \Delta_E = \bar{\Delta}_j. \tag{12}$$

The result (3) can be generalized to include the case an anisotropic film described in terms of the principal axes by the components σ_{xe} and σ_{ye} of the effective conductivity tensor. We assume as before that the local conductivity $\sigma(\mathbf{r})$ is isotropic, so that the tensor character of $\hat{\sigma}_e$ is due to the shapes and disposition of the inclusions. The derivation of the reciprocity relations is similar to the previous one. As a result we obtain two equations:

$$\sigma_{xe} \mathfrak{F}_{ye} = \sigma_1 \sigma_2, \quad \sigma_{ye} \mathfrak{F}_{xe} = \sigma_1 \sigma_2. \tag{13}$$

For systems that coincide with their reciprocals we have $\hat{\sigma}_e = \hat{\sigma}_e$ and Eqs. (13) degenerate into a single one:

$$\sigma_{xe}\sigma_{ye}=\sigma_{i}\sigma_{2}, \qquad (14)$$

so that it is impossible to determine both components of the effective conductivity tensor. Nonetheless relations (13) can be useful for different calculations.

3. A two-dimensional isotropic system in an external transverse magnetic field is described by the conductivity tensor

$$\delta = \begin{pmatrix} \sigma_x & \sigma_a \\ -\sigma_a & \sigma_x \end{pmatrix}.$$
 (15)

The galvanomagnetic properties of two-dimensional two-component systems were investigated by Dykhne² (see also Refs. 3 and 4). The reciprocity relations can be obtained by the same method as above, and coincide with Eq. (4) of the preceding paper⁴ with the following substitutions:

 $\sigma \rightarrow \sigma_e, \sigma' \rightarrow \tilde{\sigma}_e, \beta \rightarrow \beta_e, \beta' \rightarrow -\tilde{\beta_e}.$

Expressing the quantities in these equations in terms of of the components of the tensor (15) we obtain two reciprocity relations:

$$\bar{\sigma}_{se} = \sigma_{se} \frac{1 + b'd'}{\sigma_{se}^2 d'^2 + (\sigma_{se} d' - 1)^2},$$

$$\bar{\sigma}_{ee} = \frac{(\sigma_{se}^2 + \sigma_{ae}^2)d' - \sigma_{se}(1 - b'd') - b'}{\sigma_{re}^2 d'^2 + (\sigma_{se} d' - 1)^2}.$$
(16)

Here

$$b' = \frac{\sigma_{x2}(\sigma_{x1}^{2} + \sigma_{a1}^{2}) - \sigma_{x1}(\sigma_{x2}^{2} + \sigma_{a2}^{2})}{\sigma_{x1}\sigma_{a2} - \sigma_{x2}\sigma_{a1}},$$

$$d' = \frac{\sigma_{x1} - \sigma_{x2}}{\sigma_{x1}\sigma_{a2} - \sigma_{x2}\sigma_{a1}}.$$
 (17)

The reciprocal system is obtained from the initial one by the substitution $(\sigma_{x1}, \sigma_{a1}) = (\sigma_{x2}, \sigma_{a2})$.

In addition, as shown by Dykhne,² the components of the effective conductivity tensor are connected by an exact relation [see Eqs. (7) and (6) of Ref. 4] that is valid for arbitrary concentrations, shapes, and distribution of the inclusions

$$(\sigma_{xe}^{2}+\sigma_{ae}^{2})d-\sigma_{ae}-b=0, \qquad (18)$$

where

$$b = \frac{\sigma_{a_1}(\sigma_{a_2}^2 + \sigma_{a_2}^2) - \sigma_{a_2}(\sigma_{x_1}^2 + \sigma_{a_1}^2)}{(\sigma_{x_1}^2 + \sigma_{a_1}^2) - (\sigma_{x_2}^2 + \sigma_{a_2}^2)},$$

$$d = (\sigma_{a_1} - \sigma_{a_2}) / [(\sigma_{x_1}^2 + \sigma_{a_1}^2) - (\sigma_{x_2}^2 + \sigma_{a_2}^2)].$$
 (19)

From (18) and (19) (with the substitution $\hat{\sigma}_1 = \hat{\sigma}_2$) it is seen that the reciprocal system also satisfies Eq. (18), so that relations (16) are not independent. This can be verified by directly substituting (16) in the equation obtained from (18) by the substitution $\hat{\sigma}_1 = \hat{\sigma}_2$. In the corresponding calculations it is convenient to use the readily verified identity

b'd+bd'+1=0.

If the reciprocal system is converted by the additional substitution p-1-p into the initial one, then

$$\tilde{\sigma}_{xe}(1-p) = \sigma_{xe}(p), \quad \tilde{\sigma}_{ae}(1-p) = \sigma_{ae}(p). \tag{20}$$

The conditions (20), just as (7), are satisfied for a limited class of systems. In particular, they are valid for a randomly inhomogeneous medium and for a system of the checkerboard type, for which we obtain from (16)-(20) at $p = \frac{1}{2}$

$$\sigma_{xe} = (\sigma_{x1}\sigma_{x2})^{\frac{1}{2}} \left[1 + \left(\frac{\sigma_{a1} - \sigma_{a2}}{\sigma_{x1} + \sigma_{x2}} \right)^2 \right]^{\frac{1}{2}}$$

$$\sigma_{ae} = \frac{\sigma_{x1}\sigma_{a2} + \sigma_{x2}\sigma_{a1}}{\sigma_{x1} + \sigma_{x2}}, \quad \tau = 0.$$
(21)

Expressions (21) generalize somewhat the results of Dykhne² and can be obtained also from the corresponding equations of the preceding paper.⁴

At $p = \frac{1}{2}$ relations (16)-(20) yield the components of the effective conductivity tensor at $p < p_c$ if they are known at $p > p_c$ (and vice versa). Moreover, for a complete description of the galvanomagnetic properties of such systems in the entire concentration region it suffices to know only one of the components of the tensor $\hat{\sigma}_e$ at all $p \ge p_c$ (or $p \le p_c$).

We now use (16) and (18) to determine the galvanomagnetic properties of two-dimensional two-component media. We consider first for this purpose a system in the form of a conductivity matrix $\hat{\sigma}_2$ with ideally conducting (superconducting) inclusions $(\hat{\sigma}_1 + \infty)$ whose concentration is less than critical $(p < p_c, \tau < 0)$. In this case the electric field E inside the inclusions is zero, so that the boundary condition is the vanishing of the tangential component of the field, $E_t = 0$. We introduce in place of the current density j the vector

$$\mathbf{J} = \mathbf{j} - \hat{\mathbf{\sigma}}_{a^2} \mathbf{E},\tag{22}$$

where $\hat{\sigma}_{a2}$ denotes the off-diagonal part of the conductivity tensor $\hat{\sigma}_{2}$. The dc equations outside the inclusion then become

div J=0, rot E=0,
$$J=\sigma_{x2}E$$
 (23)

with the boundary condition $E_t = 0$. The problem is thus completely analogous to that of calculating the conductivity $\sigma_e(\tau < 0)$ of a system with superconducting inclusions in a zero magnetic field, so that

$$\sigma_{xe} = \sigma_{x2} f_s, \quad \tau < 0, \tag{24}$$

here the function $f_s = f_s(\tau)$ determines the dependence of $\sigma_s(\tau < 0)$ on τ :

$$\sigma_{\mathfrak{s}}(\tau < 0) = \sigma_2 f_{\mathfrak{s}}(\tau).$$

In particular, $f_{s} \sim |\tau|^{-q}$ as $\tau \to 0.^{3,6}$ On the other hand, the entire dependence of σ_{xe} on the magnetic field is contained in σ_{x2} .

The expression for $\sigma_{a_{\theta}}$ can be obtained from (18) and (19) by taking the limit as $\hat{\sigma}_1 - \infty$. As a result

$$\sigma_{ae} = \sigma_{a2}, \quad \tau < 0, \tag{25}$$

which seems natural, since the quantity σ_a has in fact dropped out of the problem.

The reciprocity relations enable us now to determine the galvanomagnetic properties of the system in the form of a conductivity matrix $\hat{\sigma}_1$ with insulator inclusions $(\hat{\sigma}_2 \rightarrow 0)$, when the concentration of the conducting component exceeds critical $(p > p_c, \tau > 0)$. Choosing a reciprocal structure with superconducting inclusions and using the inequality $\hat{\sigma}_2 \ll \hat{\sigma}_1$, we obtain from (16), (17), (14), and (25)

$$\frac{\sigma_{xe}^2 + \sigma_{ae}^2}{\sigma_{xe}} = \frac{\sigma_{xi}^2 + \sigma_{ai}^2}{\sigma_{xi}} f_s^{-1}, \quad \frac{\sigma_{xe}^2 + \sigma_{ae}^2}{\sigma_{ae}} = \frac{\sigma_{xi}^2 + \sigma_{ai}^2}{\sigma_{ai}}.$$
 (26)

The second equation in (26) also follows from relation (18) as $\hat{\sigma}_2 \neq 0$. From (26) we obtain the components of the effective conductivity tensor for a system with insulator inclusions:

$$\sigma_{ze} = \sigma_{zi} \frac{(\sigma_{ai}^2 + \sigma_{zi}^2) f_d^{-1}}{\sigma_{ai}^2 + \sigma_{zi}^2 f_d^{-2}}, \quad \sigma_{ae} = \sigma_{ai} \frac{\sigma_{ai}^2 + \sigma_{zi}^2}{\sigma_{ai}^2 + \sigma_{zi}^2 f_d^{-2}}; \quad \tau > 0.$$
(27)

In (27) we have included the function $f_d = f_d(\tau)$, which describes the dependence of the conductivity of the considered system on τ at H = 0: $\sigma_e(\tau > 0) = \sigma_1 f_d(\tau)$. The functions f_d and f_s are connected, according to (3), by the relation

$$f_s(\tau)f_d(\tau) = 1. \tag{28}$$

Expressions (24), (25), and (27) were obtained for systems with "superconducting" and insulator inclusions. Outside the smearing region,^{3,6} however, they are valid also for a medium with finite but strongly differing conductivity components, $\hat{\sigma}_2 \ll \hat{\sigma}_1$.

4. In thermoelectric, thermodiffusion, and diffusionelectric and other phenomena,^{7,9} the medium is characterized by a set of currents j_i and fields E_i (i = 1, ..., n)satisfying in the linear case the equations

$$rot E_i = 0, div j_i = 0.$$
 (29)

For an isotropic medium in the absence of an external magnetic field, the linear connection between \mathbf{j}_1 and \mathbf{E}_k is given by the "Ohm's law"

$$\mathbf{j}_i = \sigma_{ik} \mathbf{E}_k, \tag{30}$$

where the matrix of the kinetic coefficients $\hat{\sigma}$ in the

inhomogeneous system depends on the coordinates. As usual, summation over the repeated indices is implied in (30) and hereafter (i, k = 1, ..., n).

In a two-dimensional two-component system it is also possible to obtain reciprocity relations for the matrix $\hat{\sigma}_{o}$ of the effective kinetic coefficients. To find them we generalize (1) by transforming to a primed frame

$$\mathbf{j}_{i} = \lambda_{ik} [\mathbf{n} \times \mathbf{E}_{k'}], \quad \mathbf{E}_{i} = \mu_{ik} [\mathbf{n} \times \mathbf{j}_{k'}]. \tag{31}$$

The matrices $\hat{\lambda}$ and $\hat{\mu}$ do not depend on the coordinates. The equations for the currents and the fields in the primed system are of the form (29) and (39) with a kinetic-coefficient matrix

$$\hat{\sigma}'(\mathbf{r}) = \hat{\mu}^{-1} \hat{\sigma}^{-1}(\mathbf{r}) \hat{\lambda}.$$
(32)

The reciprocity relations are found as above, and coincide in form with (32)

$$\hat{\sigma}_{e}\hat{\mu}\sigma_{e}=\hat{\lambda}.$$
 (33)

To calculate the matrices $\hat{\lambda}$ and $\hat{\mu}$ we impose the condition that the primed system is the reciprocal one and differs from the initial one by the substitution $\hat{\sigma}_1 \neq \hat{\sigma}_2$, where $\hat{\sigma}_1$ and $\hat{\sigma}_2$ are the values of the matrix $\hat{\sigma}$ in the first and second components. This yields equations for the unknown matrices $\hat{\lambda}$ and $\hat{\mu}$:

$$\hat{\sigma}_{i} = \hat{\mu}^{-i} \hat{\sigma}_{2}^{-i} \hat{\lambda}, \quad \hat{\sigma}_{2} = \hat{\mu}^{-i} \hat{\sigma}_{i}^{-i} \hat{\lambda}. \tag{34}$$

The equalities in (34) constitute a linear homogeneous system of equations in the components of the matrices $\hat{\lambda}$ and $\hat{\mu}$. We determine the conditions for solvability of this system after first eliminating from (34) the matrix $\hat{\lambda}$:

$$\hat{\sigma}_{i}\hat{\mu}\hat{\sigma}_{2}-\hat{\sigma}_{2}\hat{\mu}\hat{\sigma}_{i}=0. \tag{35}$$

The system solvability condition (35) can in general impose certain restrictions on the matrices $\hat{\sigma}_1$ and $\hat{\sigma}_2$.

If the current densities \mathbf{j}_1 and the corresponding fields \mathbf{E}_i are conjugate,¹⁰ then according to the Onsager principle of the symmetry of the kinetic coefficients¹⁰ the matrix $\hat{\sigma}$ is symmetric. In this case, with the matrix $\hat{\mu}$ symmetric the condition (35) for the solvability of the system is satisfied. Moreover, for the n(n+1)/2 un-

knowns the system (35) contains n(n-1)/2 equations, so that n-1 components of the matrix $\hat{\mu}$ can be choosen arbitrary, say equal to zero. In particular, at n=2, putting $\mu_{12}=0$, we arrive at the conclusion that the matrix $\hat{\mu}$ can be sought in diagonal form. In the case of asymmetrical matrix $\hat{\sigma}$, the investigation of the system solvability condition (35) in general form is difficult. In the case n=2 of practical interest, however, direct calculations show that the determinant (35) of the system vanishes identically, so that the solvability condition is satisfied.

For systems that coincide at $p = \frac{1}{2}$ with their reciprocals $(\hat{\sigma}_e = \hat{\sigma}_e)$, we can obtain from (33) all the components of the tensor $\hat{\sigma}_e$. In particular, for the effective thermoelectric coefficient α_e we obtain the simple expression

$$\alpha_{e} = \frac{\alpha_{1}(\sigma_{1}\kappa_{2})^{\nu_{h}} + \alpha_{2}(\sigma_{2}\kappa_{1})^{\nu_{h}}}{(\sigma_{1}\kappa_{2})^{\nu_{h}} + (\sigma_{2}\kappa_{1})^{\nu_{h}}},$$
(36)

where α_i , σ_i , and \varkappa_i are respectively the thermoelectric coefficient, the conductivity, and the thermal conductivity of the *i*-th component.

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